



VU Research Portal

Continuous wave coherent Lyman-alpha radiation

Eikema, K.S.E.; Walz, J.; Hansch, T.W.

published in

Physical Review Letters

1999

DOI (link to publisher)

[10.1103/PhysRevLett.83.3828](https://doi.org/10.1103/PhysRevLett.83.3828)

document version

Publisher's PDF, also known as Version of record

[Link to publication in VU Research Portal](#)

citation for published version (APA)

Eikema, K. S. E., Walz, J., & Hansch, T. W. (1999). Continuous wave coherent Lyman-alpha radiation. *Physical Review Letters*, 83, 3828-3831. <https://doi.org/10.1103/PhysRevLett.83.3828>

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal ?

Take down policy

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

E-mail address:

vuresearchportal.ub@vu.nl

Continuous Wave Coherent Lyman- α Radiation

K. S. E. Eikema, J. Walz, and T. W. Hänsch

Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Strasse 1, 85748 Garching, Germany

(Received 7 July 1999)

We describe the first coherent generation of continuous wave radiation at 121.56 nm. A yield of 3×10^8 photons/sec (0.5 nW) has been achieved by four-wave mixing in mercury vapor. This continuous Lyman- α source could be used for laser cooling and precision spectroscopy of both hydrogen and antihydrogen.

PACS numbers: 42.65.Ky, 32.80.Pj

The hydrogen atom is an important testing ground for fundamental theories. High resolution laser spectroscopy has provided cornerstones such as the measurement of fundamental constants, a test of quantum electrodynamics, and even the investigation of hadronic structure [1]. The recent production of antihydrogen atoms [2], albeit at almost the speed of light, has given prospect of a new field. Precise tests of the fundamental *CPT* symmetry [3] and even a comparison of the gravitational force on matter and antimatter [4] now challenge both experimental and theoretical physicists. Two experiments using the new Antiproton Decelerator (AD) at CERN [5] are now in preparation with the goal of precision measurements on magnetically trapped antihydrogen [6].

The narrow $1S$ - $2S$ transition is an especially intriguing candidate for ultimate precision experiments. The excitation rate for this two-photon transition, however, is typically rather small. Previous experiments on ordinary hydrogen compensated that by the use of many atoms (10^{15} – 10^{17} atoms/s in a beam [7,8], 10^{10} – 10^{13} atoms in a trap [9]). Given that the AD will deliver about 10^7 antiprotons/min at 100 MeV/c [5], it is clear that new techniques need to be developed for experiments with antihydrogen.

In this Letter we report the first production of continuous coherent radiation at 121.56 nm. Radiation at this wavelength of Lyman- α can be used to excite the $1S$ - $2P$ single photon transition, which is the strongest resonance in the (anti-)hydrogen atom, and the only one that allows for efficient laser cooling.

Many pulsed Lyman- α sources have been built over the years (see, e.g., [10–14]). Using such a pulsed source (2×10^9 photons per 10 ns pulse at a repetition rate of 50 Hz), it has even been possible to demonstrate laser cooling of magnetically trapped hydrogen to 8 mK [15]. A *continuous* Lyman- α source, however, has distinct advantages over pulsed sources. Its much narrower spectral bandwidth provides higher selectivity for magnetic substrates of atoms in a trap. This reduces losses due to spurious optical pumping to untrapped magnetic sublevels. Laser cooling and Zeeman slowing can be more efficient, if they are not hampered by the low duty cycle from pulsed high-power lasers. Even high resolution

spectroscopy of the weak $1S$ - $2S$ transition with only a few atoms seems possible with the help of continuous Lyman- α radiation on the strong $1S$ - $2P$ transition using a “shelving” scheme [16].

Our Lyman- α source is based on continuous four-wave mixing (CW-FWM) in mercury vapor. An important requirement for CW-FWM is resonant enhancement of the nonlinear susceptibility given the much lower power levels available from continuous lasers than from pulsed lasers. Therefore an exact two-photon resonance must be utilized, in combination with near one- and three-photon resonances. Generation of continuous vacuum ultraviolet (VUV) in this fashion was first reported by Freeman *et al.* [17]. Subsequent experiments demonstrated the feasibility of generating VUV down to 133 nm (11 pW) [18–20].

For Lyman- α generation mercury is well suited because of its unusual high ionization limit of $84\,184\text{ cm}^{-1}$. As a result bound resonances such as the $12p\ ^1P_1$ and the $11p\ ^1P_1$ near Lyman- α contribute significantly to the nonlinear susceptibility [21]. We have investigated two different four-wave-mixing schemes in mercury that are depicted in Fig. 1. One scheme uses light fields at 257 nm and at 399 nm to establish a two-photon resonance with the $7s\ ^1S_0$ state. The wavelength of the third incident light field is chosen such that the sum frequency is at Lyman- α . The second scheme uses two photons from a light field at 280 nm to establish a two-photon resonance with either the $6d\ ^1D_2$ or the $6d\ ^3D_2$ state. From an experimental point of view the second scheme is much easier, because it needs only two different colors. However, it is also less efficient as will be discussed later.

The experimental setup for the first scheme ($7s\ ^1S_0$ two-photon enhancement) is shown in Fig. 2. The first fundamental wavelength at 257 nm is obtained from a frequency doubled single mode Ar⁺ laser (Spectra-Physics 2030). From 2.2 W at 514.5 nm, typically 750–800 mW is generated at 257 nm in β -barium borate (BBO, from Cleveland crystals). Flushing the crystal with pure oxygen during operation and heating to 50 °C was necessary to prevent optical damage to the crystal. All frequency doubling was performed with z -fold 4-mirror enhancement resonators, locked by the Hänsch-Couillaud technique [22].

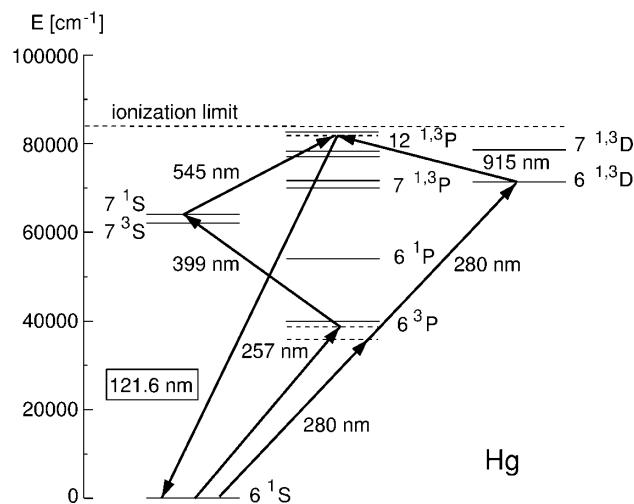


FIG. 1. Simplified level scheme of mercury. The arrows indicate the two four-wave-mixing schemes investigated.

The second step at 399 nm is realized by frequency doubling a single mode Ti:sapphire laser (Coherent 899) in lithium borate (LBO, from Photox). In this case 1.2 W fundamental power at 799 nm results in ≈ 300 mW at 399 nm. The LBO crystal is also heated to 50 °C, and flushed with dry nitrogen to keep the optical surfaces free from dust. The third wavelength required is obtained from a frequency stabilized dye laser (Coherent 699) operating on rhodamine 110. By scanning this laser from 543 to 584 nm a VUV range between 121.4 and 123.4 nm is covered. Astigmatism and ellipticity of the frequency doubled beams are compensated for by cylindrical lenses. Telescopes with spatial filtering enlarge the beams to match focusing parameters. The three fundamental beams of parallel and linear polarization are then focused ($f = 20$ cm, confocal parameters $b \approx 0.18$ – 0.22 cm) in the middle of a ≈ 5 cm long mercury vapor zone produced by a conical heat-pipe oven. The typical mercury vapor pressure in the center of this heat pipe is set between 10 and 45 mbar, which has been deduced from temperature measurements (180–220 °C). Additional heating elements and 40 mbar of helium buffer gas prevent condensation of mercury on the entrance and exit optics. The generated VUV and fundamental beams are separated with the help of the dispersion of an $f = 15$ cm MgF_2 lens and 3 mm small mirror, in combination with three narrow band VUV interference filters (Acton). The small mirror reflects the fundamental beams, but passes 50% of the Lyman- α . The interference filters have a bandwidth of 15 nm and a measured individual peak transmission of 17% at Lyman- α . A solar-blind photomultiplier (Hamamatsu R1459) is used for counting the Lyman- α photons. We estimate that the overall detection efficiency of Lyman- α is 0.015% due to the lens ($\approx 40\%$), the small mirror ($\approx 50\%$), three filters (each $\approx 17\%$), and the photomultiplier (15%). The frequencies of the Ti:sapphire, single-mode argon-ion, and dye-laser

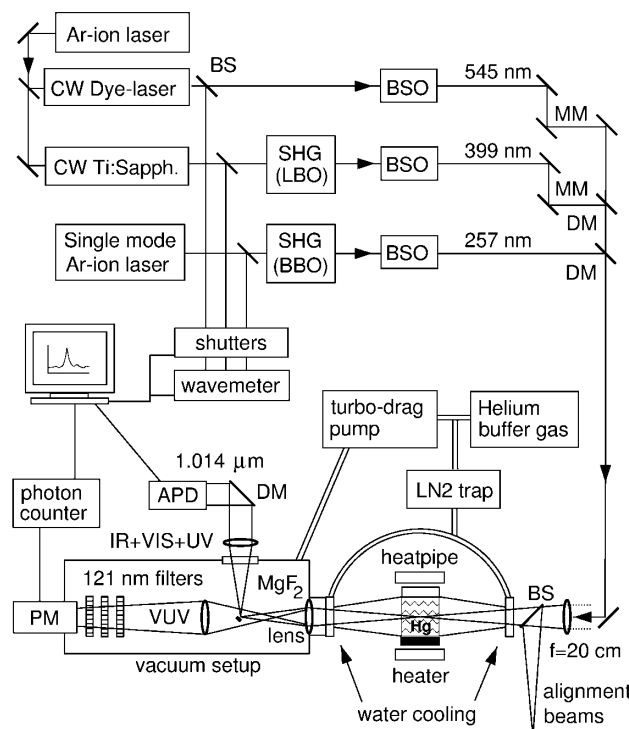


FIG. 2. Schematic of the setup for $7s^1S_0$ two-photon enhanced continuous four-wave mixing. In this figure, SHG stands for second-harmonic generation, BSO for beam shaping optics (cylindrical lenses, telescopes with spatial filtering, polarization rotation), MM for motorized mirror mounts, DM for dichroic mirror, BS for beam splitter, APD for avalanche photodiode, and LN2 for liquid nitrogen.

beams were measured with a wave meter. The VUV wavelength was determined by adding the frequencies of the fundamental beams.

Alignment of the three beam foci was found to be very critical. The overlap was preadjusted using alignment beams (see Fig. 2) and a pinhole. The 257 and the 399 nm foci were aligned further by maximizing fluorescence at $1.014 \mu\text{m}$ which originates from the decay of the two-photon resonant $7s^1S_0$ to the $6p^1P_1$ level. The 545 nm focus was then aligned on the VUV signal itself.

In Fig. 3 the VUV yield for $7s^1S_0$ enhanced FWM is given over the wavelength range that can be covered by scanning the Rhodamine 110 dye-laser (at 37 mbar mercury). These scans were performed at moderate laser power levels (effectively ≈ 77 mW at 257 nm, 67 mW at 399 nm, and maximum 0.64 W for the dye laser in the heat pipe). For increasing mercury pressures the yield at Lyman- α was found to rise faster than close to the bound resonances. In 37 mbar Hg, and with full laser power, a maximum Lyman- α count rate of 50 000 counts/s has been achieved, equivalent to 3×10^8 photons/s (0.5 nW). A maximum VUV yield of 3 nW was achieved near 121.9 nm. In this case the effective fundamental laser power in the heat pipe was 440 mW at 257 nm, 200 mW at 399 nm, and 520 mW at 545 nm. At these high

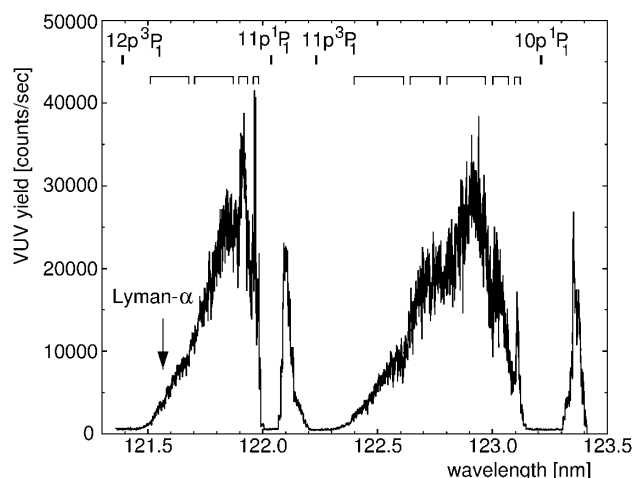


FIG. 3. Continuous four-wave-mixing yield for the $7s\ ^1S_0$ two-photon enhanced scheme, as a function of the VUV wavelength (see text for experimental conditions).

fundamental UV intensities, however, the VUV yield degraded by nearly 2 orders of magnitude in one hour. Inspection of the separation lens, after the $7s\ ^1S_0$ enhanced measurements had been completed, revealed a brownish spot where the light beams had been, mainly on the vacuum side ($<10^{-5}$ mbar). The contamination is most likely due to UV breakdown of organic molecules, as it could be removed with acetone and sticky-polymer (Opticlean) cleaning. Additional cryopumping or a buffer gas in all Lyman- α guiding sections might reduce this problem in future experiments. Power saturation is not yet expected at the intensities used. However, the VUV yield at full UV laser power was 40% lower than expected based on the low laser power results presented above. We ascribe this mainly to a worse and more instable mode profile of the 257 nm beam due to Ar^+ pump laser instabilities. Also the rapid contamination of the separation lens for high UV power probably reduced the measured VUV yield. The bandwidth of the generated Lyman- α is expected to be a few MHz, mostly determined by the unstabilized Ar^+ laser used to produce radiation at 257 nm.

Natural mercury contains several isotopes, which influences the VUV yield. The isotope shifts are several GHz in the $7s\ ^1S_0$ state, hence only one isotope contributes to the VUV enhancement at the time. The ^{202}Hg isotope is the most abundant (30%), and has therefore been used (put on resonance) for all the measurements. Given that the density enters the yield quadratically, an order of magnitude more Lyman- α is expected from an isotopically pure mercury vapor.

The second scheme for CW-FWM we investigated is based on a two-photon resonance with either the $6d\ ^1D_2$ or the $6d\ ^3D_2$ state (see Fig. 1). These states are only 63 cm^{-1} apart. The setup for this scheme is in principle similar to the one previously discussed, except for the different wavelengths used. In this case the required 280 nm

for the two-photon resonant step is produced by frequency doubling the rhodamine 110 dye laser in a 5 cm long potassium dihydrogen phosphate crystal (Cleveland Crystals). From 1.15 W fundamental power at 560 nm, typically 230–265 mW at 280 nm is produced. The Ti:sapphire supplied the required infrared near 946–873 nm to reach VUV wavelengths around Lyman- α (700–900 mW). In Fig. 4 the VUV production near Lyman- α is shown for 34 mbar of mercury. To cover the required wavelength range the Ti:sapphire laser was scanned by rotating the internal birefringent filter. Only for the measurement shown in Fig. 4(A), this resulted in spurious structure by reproducible infrared intensity fluctuations of up to 30%. Therefore this measurement was divided by the (globally almost flat) normalized infrared intensity. The effective 280 nm and IR laser power in the heat pipe were 180 mW and 500 mW, respectively. No degradation over time of the VUV production was seen at those power levels. The maximum VUV intensity is generated by the $6d\ ^1D_2$ enhanced scheme, but for Lyman- α the $6d\ ^3D_2$ enhanced scheme is more efficient. In this case a maximum Lyman- α yield of 1.9×10^6 photons/s has been achieved (from effectively 230 mW at 280 nm and 500 mW at 919 nm in the heat pipe). Compared to $7s\ ^1S_0$ enhanced FWM, however, the aforementioned schemes are at least 2 orders of magnitude less effective in producing Lyman- α . The lower yield is in part due to the larger detuning from the $6p\ ^3P_1$ state, the lower fundamental power product, and a mismatch of confocal parameters of UV ($b = 0.21\text{ cm}$) and IR beam ($b = 0.63\text{ cm}$) due to the large fundamental wavelength difference.

Shorter VUV wavelengths have been generated by tuning the Ti:sapphire laser from 873 to 774 nm. In this manner continuous VUV from 120.7 down to 119 nm could be generated as shown in Fig. 5. The generated 119 nm is the shortest wavelength so far reported for CW-FWM.

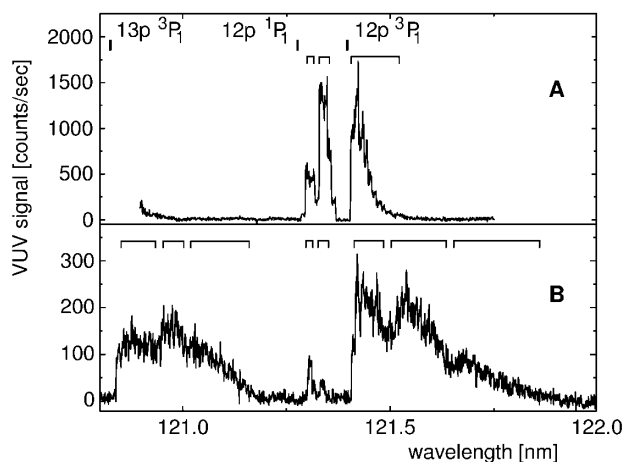


FIG. 4. Generation of light near Lyman- α for two-photon enhancement by the $6d\ ^1D_2$ (A) and the $6d\ ^3D_2$ state (B). The VUV signal in (A) was divided by the normalized infrared intensity (see text).

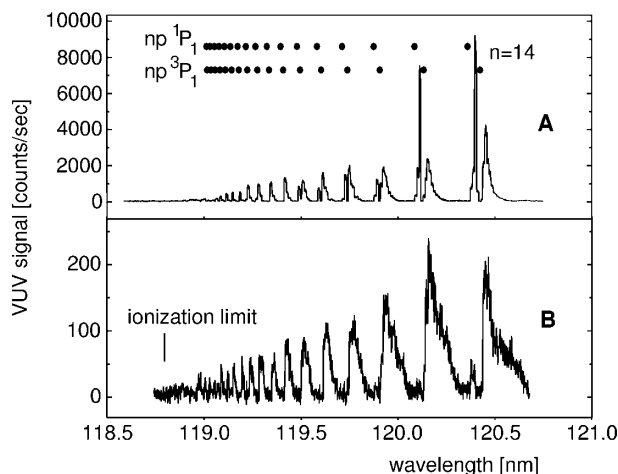


FIG. 5. Continuous four-wave-mixing yield three-photon enhanced by the $n^{1,3}P_1$ Rydberg series. Two-photon enhancement by the $6d^1D_2$ (A) and the $6d^3D_2$ state (B).

An interesting feature of the measured VUV yield curves in Figs. 3 and 4 is a reproducible oscillatory structure in the global yield curves with a varying amplitude, and a period ranging from roughly 0.2 to 0.01 nm. The approximate period lengths and positions are marked in both figures by horizontal bars directly above the VUV-yield curves. For lower density the yield oscillations reduced or disappeared. Although other causes cannot be excluded entirely, theoretical calculations show that the oscillations can be explained to be a result of strong absorption of the generated wave and a wavelength dependent phase mismatch. Theoretically the strongest VUV yield oscillations appear for a wavelength independent absorption coefficient in the range of $5\text{--}15\text{ cm}^{-1}$ given the experimental conditions in this experiment. Absorption due to bound resonances changes too rapidly with wavelength to lead to oscillatory structures by itself. Although global absorption can explain at least the appearance of the yield oscillations, we found that the VUV absorption is probably not a bulk effect but is limited to the focal region. This can be concluded from experiments performed for the $6d$ enhanced schemes, where the mercury vapor zone was made twice as long (from ≈ 5 to ≈ 10 cm). The VUV production reduced only by a factor of ≈ 2 outside the immediate neighborhood of the $12p^{1,3}P_1$ and $13p^{1,3}P_1$ resonances. Based on the theoretical absorption coefficients, however, a reduction by several orders of magnitude is expected. If absorption is indeed the cause for the oscillations, we suspect that a substance such as mercury dimers might be produced in the focus by photoassociation. Mercury dimers have a fairly flat absorption cross section over the measured wavelength region [23]. A more in-depth investigation of VUV absorption effects and its theoretical treatment will be given in a subsequent publication.

In conclusion we have demonstrated the first generation of continuous Lyman- α radiation with a power of 0.5 nW. This opens up a new field of continuous laser cooling and sensitive spectroscopy of antihydrogen. Strong absorption of the VUV has been identified as the most probable cause for the oscillations observed. Improvements of the heat-pipe design to circumvent this effect, and other measures have the potential to improve the Lyman- α production by several orders of magnitude. Most promising in this respect is the use of electromagnetically induced transparency [24] involving the $6p^3P_1$ state. Other possibilities include an enhancement cavity for the fundamental waves [20], or the use of single isotope mercury.

It is a pleasure to thank Wim Hogervorst for fruitful discussions about four-wave mixing in mercury.

- [1] C. Schwob *et al.*, Phys. Rev. Lett. **82**, 4960 (1999); A. Huber *et al.*, Phys. Rev. Lett. **80**, 468 (1998); Th. Udem *et al.*, Phys. Rev. Lett. **79**, 2646 (1997).
- [2] G. Baur *et al.*, Phys. Lett. B **368**, 251 (1996); G. Blanford *et al.*, Phys. Rev. Lett. **80**, 3037 (1998).
- [3] R. Bluhm, V. A. Kostelecký, and N. Russell, Phys. Rev. Lett. **82**, 2254 (1999).
- [4] J. S. Bell in *Fundamental Symmetries*, edited by P. Bloch, P. Pavlopoulos, and R. Klapisch (Plenum, New York, 1987), p. 1.
- [5] S. Maury, Hyperfine Interact. **109**, 43 (1997).
- [6] See, e.g., J. Eades and F. J. Hartmann, Rev. Mod. Phys. **71**, 373 (1999).
- [7] A. Huber *et al.*, Phys. Rev. A **59**, 1844 (1999).
- [8] F. Schmidt-Kaler *et al.*, Phys. Rev. A **51**, 2789 (1995).
- [9] C. L. Cesar *et al.*, Phys. Rev. Lett. **77**, 255 (1996).
- [10] R. Mahon, T. J. McIlrath, and D. W. Koopman, Appl. Phys. Lett. **33**, 305 (1978).
- [11] R. Hilbig and R. Wallenstein, IEEE J. Quantum Electron. **19**, 1759 (1983).
- [12] J. P. Marangos *et al.*, J. Opt. Soc. Am. B **7**, 1254 (1990).
- [13] W. D. Phillips *et al.*, Hyperfine Interact. **76**, 265 (1993).
- [14] S. A. Meyer and G. W. Faris, Opt. Lett. **23**, 204 (1998).
- [15] I. D. Setija *et al.*, Phys. Rev. Lett. **70**, 2257 (1993).
- [16] H. G. Dehmelt, Bull. Am. Phys. Soc. **20**, 60 (1975).
- [17] R. R. Freeman *et al.*, Appl. Phys. Lett. **33**, 739 (1978).
- [18] J. Nolting *et al.*, Appl. Phys. B **50**, 331 (1990).
- [19] A. Timmermann and R. Wallenstein, Opt. Lett. **8**, 517 (1983).
- [20] J. Nolting and R. Wallenstein, Opt. Commun. **79**, 437 (1990).
- [21] A. V. Smith and W. J. Alford, J. Opt. Soc. Am. B **4**, 1765 (1987), and references therein.
- [22] T. W. Hänsch and B. Couillaud, Opt. Commun. **35**, 441 (1980).
- [23] S. H. Linn *et al.*, Chem. Phys. Lett. **105**, 645 (1984).
- [24] S. E. Harris *et al.*, Phys. Rev. Lett. **64**, 1107 (1990).